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PM, carbon, and PAH emissions from a diesel generator fuelled with soy-biodiesel blends

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ABSTRACT

Biodiesels have received increasing attention as alternative fuels for diesel engines and generators. This study investigates the emissions of particulate matter (PM), total carbon (TC), e.g., organic/elemental carbons, and polycyclic aromatic hydrocarbons (PAHs) from a diesel generator fuelled with soy-biodiesel blends. Among the tested diesel blends (B0, B10 (10 vol% soy-biodiesel), B20, and B50), B20 exhibited the lowest PM emission concentration despite the loads (except the 5 kW case), whereas B10 displayed lower PM emission factors when operating at 0 and 10 kW than the other fuel blends. The emission concentrations or factors of EC, OC, and TC were the lowest when B10 or B20 was used regardless of the loading. Under all tested loads, the average concentrations of total-PAHs emitted from the generator using the B10 and B20 were (by 38% and 28%, respectively) than those using pure petroleum diesel fuel (B0), while the emission factors of total-PAHs decreased with an increasing ratio of biodiesel to premium diesel. With an increasing loading, although the brake specific fuel consumption decreased, the energy efficiency increased despite the bio/petroleum diesel ratio. Therefore, soy-biodiesel is promising for use as an alternative fuel for diesel generators to increase energy efficiency and reduce the PM, carbon, and PAH emissions.

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1. Introduction

Diesel-powered engines are extensively adopted in large buses, heavy-duty trucks, construction machines, and generators due to their high fuel efficiency, high power output, and enhanced fuel savings [1,2]. However, diesel vehicles are the major source of ambient aerosols in metropolitan areas [3,4]. Durbin et al. [5] cited elemental and organic carbons as the primary constituents (73–83% of total mass) of diesel particulate matter (DPM). DPM may adversely impact the environment in different ways due to their optical, physical, chemical, and toxicological characteristics. Experimental animal studies found soot to be carcinogenic in [6], while EC and OC might induce respiratory and cardiovascular diseases, even carcinoma [7–11]. Several prominent national organizations, including the National Institute for Occupational Safety and Health (NIOSH) [12], the International Agency for Research on Cancer (IARC) [13], the World Health Organization (WHO) [14], the U.S. Environmental Protection Agency [15], and the U.S. National Toxicology Program [16], have classified diesel exhaust as a likely human carcinogen. Diesel vehicles also emit toxic and carcinogenic polycyclic aromatic hydrocarbons (PAHs) [17,18].

Diesel engines can be used for on-road or non-road (off-road) purposes. Despite the small number in operation as non-road diesel engines, diesel engines account for a disproportionate fraction of particulate matter (PM) and NO_x emissions because they typically have minimal emission control [19]. For non-road diesel engines in the United States, the smallest class of diesel generator (<19 kW) comprised 18% of the non-road market in the United States in 2004 [20], subsequently generating 44% of total diesel PM and 12% of NO_x emissions from mobile sources nationwide [21]. Bunger et al. [22] found that a non-road diesel engine (52 kW) emitted smaller numbers of larger particles when using biodiesel than using petroleum diesel. Liu et al. [23] monitored an 80 kW diesel generator, indicating that the DPM emissions from non-road diesel engines were significantly higher than those from on-road sources.

Using biodiesels instead of fossil diesels in (on-road and non-road) diesel engines may alleviate the emissions of carbon

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Fig. 1. Scheme of the sampling system and the generator/engine.

monoxide (CO), carbon dioxide (CO₂), total hydrocarbons (THC), sulfur dioxide (SO₂) and PAH emissions [24–34]. In contrast, some studies indicated that diesel engines fuelled with biodiesels might extend the emissions of NO_x [25,28]. Recently, diesel-engine generators have been adopted as emergency electric power in mansions [23,35–37]. Additionally, although diesel-engine generators are pervasive in some countries due to rapidly expanding industries, the expansion of electricity power has not kept pace with the continuously expanding demand in some areas [38–40].

Pollutant emissions from diesel engines depend on factors such as load, fuel type, engine type, engine maintenance, individual operator, emission control device, and lubricant oil composition. Some studies have examined OC/EC emissions under various load conditions for heavy-duty diesel vehicles [41], military vehicles [42], and non-road diesel generators [23]. However, the emission of organic/elemental carbons from non-road diesel generators fuelled with soy-biodiesel blends has seldom been studied. This study investigates the emissions of PAHs and particlebound organic/elemental carbons from a generator fuelled with efficiency are also examined at various generator loads and for different soy-biodiesel blends.

soy-biodiesel blends. Additionally, fuel consumption and energy

2. Methods and materials

2.1. Diesel generator and sample collection

This study used four soy-biodiesel blends with different mixing ratios of soy-biodiesel to premium diesel: premium diesel fuel (B0), B10 (10 vol% soy-biodiesel + 90 vol% B0), B20, and B50. The generator has a diesel engine (without catalyst installation) (NM260L, Mitsubishi) with the following specifications: one cylinder, four strokes, direct injection, water-cooled, bore and stroke of 113 mm × 115 mm, total displacement volume of 1153.3 mL, and maximum horsepower of 11.5 kW at 2600 rpm. Fig. 1 shows the sampling equipment of the generator. Tests of particle and PAH emissions were performed at loads of 0, 5, 7, and 10 kW of a diesel generator fed with four fuel types.

Parameter		Test method				
	ВО	B10	B20	B50	B100	
Cetane index	56	56	55	53	52.7 ^a	ASTM D976
Heating value (cal g ⁻¹)	11035.7 ^a	10881.3 ^b	10726.9 ^b	10263.8 ^b	9491.9 ^a	
Density at 15 °C (g cm ⁻³)	0.830	0.834	0.840	0.856	0.868 ^a	CNS 12017
C (wt%)	86.13 ^a	-	-	-	76.96ª	
H (wt%)	13.93ª	-	-	-	11.85 ^a	
O (wt%)	$\sim 0^{c}$	-	-	-	9.41 ^a	
S (ppmw)	36	32	29	18	-	ASTM D4294

(-) Not available.

Table 1

^a Data from Lin and Lin [60].

^b Calculated based on mixing ratio.

Properties of the premium diesel and soybean biodiesel.

^c Obtained by O (wt%) = 100% - C (wt%) + H (wt%) assuming that the content of other components is negligible.



Fig. 2. Emission concentrations and rates of PM from the diesel generator.

An auto-detector flow sampling system equipped with quartz fiber filters (with diameters of 47 mm) was installed downstream of the diesel generator exhaust to determine suspended particles and particulate phase PAHs. Gas phase PAHs were collected by two connected cartridges (filled with XAD-16 resins). The quartz filters were pretreated before sampling by heating in a muffle furnace in air for 2.5 h at 900 °C. Before weighing on an electronic six-digit balance ($\pm 2 \mu g$) before and after sampling, the filters were conditioned for 24 h at 25 °C and 40% relative humidity. For each sample, the concentration of suspended particulate matter was determined through means of dividing the particle mass by the volume of sampled air.

2.2. Fuel

Premium diesel fuel used in this study was obtained from the Chinese Petroleum Corporation in Taiwan. The commercial soybean biodiesel (supplied by the Word Energy, a large biodiesel producer in the United States) was produced from fresh soybean oil and methyl alcohol via a transesterification reaction. Table 1 lists the fuel properties.

2.3. Chemical analysis

The concentrations of particle-bound total and elemental carbons (TC and EC, respectively) were determined using an elemental analyzer (Carlo Erba EA 1110). Notably, different measurement methods normally lead to a variation of determined carbon content. Recently, Chow et al. [43] compared the carbon content measured using the IMPROVE and NIOSH methods. According to that study, the two methods differed mainly in the allocation of carbon evolving at 850 °C set by the NIOSH in a helium atmosphere to OC rather than to EC. This study attempted to determine the carbon content of particles by using a method (compatible OR comparable) with IMPROVE-TOR. For carbon species analysis, a weighted sample in a tin capsule was placed in an autosampler drum for deaeration. The sample was then introduced with helium into a 1000 °C vertical quartz tube to oxidize the He-carried flow and yielded a gas mixture flowing into a chromatographic column followed by a thermoconductivity detector (TCD). Next, a quarter of each sample filter was heated in a 340 °C oven for 100 min to expel the OC content, followed by feeding the sample into the element analyzer to obtain the EC content. Another quarter of each filter was fed directly into the elemental analyzer without 340 °C-heating pretreatment to guantify the TC concentration. The OC concentration was obtained from the difference between the TC and EC values. Finally, carbon content was determined using an organic analytical standard (OAS, Elemental Microanalysis Limited, B2038) containing purified urea was used as a routine working standard.

2.4. PAH analysis

Each collected sample was extracted in a Soxhlet extractor with a mixed solvent (n-hexane and dichloromethane 1:1 (v/v), 750 mL each) for 24 h. The extracts were then concentrated, cleaned up (using a silica column filled with silica gel particles (size range = 0.04–0.063 mm) positioned under a layer of anhydrous Na_2SO_4 (~1 cm high) and above a support of glass fiber), and reconcentrated by purging with ultra-pure nitrogen to exactly 1.0 mL for GC/MS analysis. The analysis method of PAHs was provided in our previous studies [33,34,44-46]. According to the molecular weights of 21 PAH compounds, the PAHs were divided into three categories: low molecular weight (LMW)-, medium molecular weight (MMW)-, and high molecular weight (HMW)-PAHs. The LMW-PAHs included naphthalene (Nap), acenaphthylene (AcPy), acenaphthene (Acp), fluorine (Flu), phenanthrene (PA), and anthracene (Ant) while the MMW-PAHs were fluoranthene (FL), pyrene (Pyr), benzo[a]anthracene (BaA), and chrysene (CHR). The HMW-PAHs were the group of cyclopenta[c,d]pyrene (CYC). benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[e]pyrene (BeP), benzo(a)pyrene (BaP), perylene (PER), dibenzo[a,h]anthracene (DBA), benzo[b]chrycene (BbC), indeno[1,2,3,-cd]pyrene (IND), benzo[ghi]perylene (Bghip), and coronene (COR). The sum of one set of 21 individual PAH data yielded a corresponding total-PAH value for the diesel generator exhaust. GC/MSD was calibrated with a diluted standard solution of 16 PAH compounds (PAH mixture-610M from Supelco, USA) plus five additional individual PAHs (from Merck, Germany). Detailed information on the GC/MSD operation, PAH quantification, QA/QC, and method detection limits can be found elsewhere [33,34,44-46].

2.5. Data analysis

For each sample, the total-PAH concentration was obtained from the sum of those of 21 PAH compounds. Additionally, the PAH homolog distribution was analyzed not only for 21-PAH, but also for molecular weight categorized PAHs (LMW-PAHs, MMW-PAHs, and HMW-PAHs). Because several PAH compounds are human carcinogens, the carcinogenic potencies of emitted PAH were determined. The carcinogenic potency of a given PAH compound was then assessed based on its benzo[a]pyrene equivalent concentration (BaP_{eq}). Using the toxic equivalent factor (TEF) of a PAH com-



Fig. 3. Emission concentrations and rates of PM-bound carbons from the generator.



Fig. 4. Carbon content of PM emitted from the diesel generator.

Table	2
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Emission concentrations and rates of PAH and BaPeq from the diesel generator under various loadings.

	Unload			5 kW			7 kW			10 kW						
	B0	B10	B20	B50	B0	B10	B20	B50	B0	B10	B20	B50	B0	B10	B20	B50
Concentrations(μ g m ⁻³) (n=3)																
LMW-PAHs	142	64.6	69.5	186	240	103	135	285	265	156	214	344	390	307	345	514
MMW-PAHs	15.8	9.89	10.7	15.8	13.1	14.2	14.5	21.4	15.7	10.8	12.4	12.8	32.4	13.3	35.4	27.5
HMW-PAHs	47.8	32.3	30.7	52.6	46.7	62.5	47.6	47.7	42.6	29.9	33.2	33.3	72.9	52.4	55.9	50.5
Total-PAHs	206	107	111	254	300	179	197	354	323	196	259	390	495	373	436	592
Total-BaP _{eq}	19.3	13.9	12.5	22.1	19.9	27.2	20.9	19.0	18.5	12.9	14.2	14.5	30.2	22.3	22.2	18.7
Emission factors $(mgL^{-1})(n=3)$																
LMW-PAHs	30.8	6.97	5.82	12.5	65.6	10.2	10.9	15.2	73.6	17.7	17.4	21.9	78.6	33.0	30.5	25.9
MMW-PAHs	3.40	1.07	0.89	1.04	3.57	1.43	1.17	1.14	4.42	1.23	1.01	0.81	6.60	1.42	3.14	1.39
HMW-PAHs	10.3	3.47	2.57	3.49	12.8	6.30	3.84	2.57	12.2	3.41	2.71	2.11	16.1	5.62	4.95	2.56
Total-PAHs	44.5	11.5	9.29	17.0	82.0	18.0	15.9	18.9	90.2	22.3	21.2	24.8	101	40.0	38.6	29.8
Total-BaP _{eq}	4.16	1.49	1.05	1.47	5.46	2.74	1.69	1.02	3.49	1.47	1.16	0.92	6.75	2.39	1.96	0.95

pound to calculate its BaP_{eq} concentration is a common practice. TEF represents the relative carcinogenic potency of a given PAH compound, in which benzo[a]pyrene is used as a reference compound to adjust its original concentration. Only a few proposals for TEFs are currently available. In this study, TEFs reported by Nisbet and LaGoy [47] were adopted. Accordingly, the carcinogenic potency of total-PAHs (i.e. total-BaP_{eq}) was assessed by summing up the BaP_{eq} concentrations estimated for each PAH compound with a corresponding TEF in the total-PAHs.

3. Results and discussion

3.1. PM emission concentrations and factors

Fig. 2 displays the emission concentrations and factors of PM from the diesel generator at various loads using diesel blends with different ratios of petroleum diesel to soybean biodiesel. Among the different generator loadings (unload (0 kW), low (5 kW), medium (7 kW), and high (10 kW)) cases, the 5 kW case had the lowest emission concentration of particulate matter (PM), when using pure petroleum diesel (B0). Dissimilarly, the PM emission concentration increased with an increasing loading if B10, B20, and B50 were used. Among the different diesel blends (B0, B10, B20, and B50), the B20 exhibited the lowest PM emission concentration despite a load (except for the 5kW case), whereas B10 exhibited lower PM emission factors when operated at 0 and 10 kW than the other fuel blends. McCormick [48] also indicated that adding 20% (the most common addition ratio) biodiesel fuel in a diesel generator facilitated the reduction of pollutants in emitted gas. The B50 had the highest PM emission concentration at all the loads; a similar trend was observed except for the 10 kW case. The mean emission factor of B50 increased by 15% over that of B0. This phenomenon probably resulted from incomplete combustion of fuel in the combustion chamber when the nebulization efficiency of nozzle was insufficient due to the increases of cetane number and viscosity after adding a significant amount of biodiesel. Lin et al. [34] indicated that although palm-biodiesel could reduce PAHs emission, the biodiesel cannot exceed 35% for PM emission control. Nevertheless, either system preheating [49,50] or fuel emulsification [45,51] can reduce the PM emission.

3.2. Effect of fuel composition on PM-bound carbon emission

The variation trends of EC emission concentrations from various fuel blends (B0 to B50) were similar at all of the tested loadings. This phenomenon is also true for those of OC and TC emissions, except for that of OC at 10 kW (Fig. 3). The emission concentrations of EC, OC, and TC were the lowest when B20 was used (with 14%, 29%, and 20% mean decreases, respectively, in comparison with those of B0)

while B50 had the highest EC, OC, and TC emission concentrations regardless of the loadings. At all of the tested loads, B10 or B20 cases exhibited the lowest emission factors of EC, OC, and TC. In comparison with B0, the use of B10 and B20 could reduce EC, OC, and TC emission factors from the diesel generator by 2–41% (average 16%), 6–48% (average 27%), and 8–42% (average 20%), respectively, regardless of the load.

The mean TC content (mass of TC/mass of PM) (TC=EC+OC) emitted from the generator were 53.0–96% while the mean EC and OC content ranged \sim 39–62% and \sim 14–37%, respectively (Fig. 4). The TC amounts positively correlated (r=0.95–0.99) with the loadings, regardless of the load and the ratio of soybean biodiesel to premium diesel fuel. Sharma et al. [52] investigated the particles emitted from a diesel generator, indicating a positive correlation between emitted EC content and generator bearing power; however, the correlation was negative if OC replaced EC. At all of the loadings (0–10 kW), the emitted EC and OC from the generator for the B10 and B20 were obviously lower than those for pure petroleum diesel (100% diesel (B0)). This phenomenon may be attributed to the higher oxygen content in biodiesel fuel than in pure petroleum diesel, leading to a more complete combustion of fuel and a decrease of carbon emissions.

Lin et al. [33] observed that the incomplete combustion of diesel fuel blends (mixed pure petroleum diesel and palm-biodiesel) occurred when the fraction of palm-biodiesel was 50% or over more, resulting in the increase in PM and PM-bound soluble organic fraction (SOF) in diesel generator exhaust. Akasaka et al. [53] indicated that the PM-bound SOF (derived partly from the lubrication oil, unburned fuel, and compounds during combustion) emission increased with the biodiesel fraction in fuel blends. Durbin et al. [5] also found that neat biodiesel had the highest total carbon emission rates for three of four test vehicles; moreover, the neat biodiesel exhibited the highest organic carbon fractions in exhaust for each of the test vehicles.

3.3. PAH emission concentrations and factors

PAHs emitted from the generator were mostly LMW-PAHs (average 76%), followed by some HMW-PAHs (average 18%) and MMW-PAHs (an average of 6%) in the lowest amount, regardless of the load and the ratio of soybean biodiesel to premium diesel fuel (Table 2). Under all the tested loadings, the average concentrations of Total-PAHs emitted from the generator using B10 and B20 were lower (by 38% and 28%, respectively) than those using the pure petroleum diesel fuel (B0). Compared with B0, the emission factors of total-PAHs were lowered (an average of 72%) as the ratio of biodiesel to premium diesel increased. The observed emission reduction is consistent with previous studies using biodiesel for on-road diesel engines [26,30,54,55].

Nevertheless, more PAH emission from the generator was observed using B50 than when using B0. PAH compounds consist of C and H elements with chemical structures of two or more fused benzene rings in linear, angular, or cluster arrangements, and can be formed in incomplete combustion or high temperature pyrolytic processes involving materials containing C and H (e.g., fossil fuels) [56]. Emissions of PAHs from combustion generally originate from three distinct mechanisms: (i) synthesis from simpler molecules in the fuel, particularly from aromatic compounds, (ii) storage in engine deposits and subsequent emission of PAHs already present in the fuel, and (iii) pyrolysis of lubricant [57]. While studying the addition of toluene in biodiesel, Kameda et al. [58] found that a slight amount of aromatic hydrocarbons in biodiesel did not significantly contribute to the reduction of PAH and nitro-PAH emissions. Similarly, Rhead and Hardy [59] also pointed out that compelling fuel manufacturers to remove aromatic compounds from fuels during refining may be less effective than anticipated if the production of aromatic compounds in diesel combustion is independent of the fuel PAH content. In this study, the soybean oil methyl ester (B100) had a higher O value (wt%) and density but a lower cetane number than pure petroleum diesel (Table 1). A moderate addition of soybean oil methyl ester in pure petroleum diesel (e.g., the B10 and B20 cases) may increase the self-ignition propensity and decrease the ignition delay of diesel, subsequently increasing fuel combustion efficiency and reducing PAH generation and emission. However, the B50 case (with a higher ratio of soybean biodiesel to petroleum diesel and larger PAH emission) is probably attributed to incomplete fuel combustion in the combustion chamber, which is associated with the poor nebulization efficiency of nozzle, due to the overdose of viscosity and the lower cetane number after adding biodiesel into petroleum diesel.

Nevertheless, Table 2 indicates that at 0, 5, 7, and 10 kW loadings, the reductions of emitted total-BaP_{eq} concentrations were 27.8%, -36.7%, 30.3%, and 26.1%, respectively, using B10 in contrast with using B0, whereas the reductions were 35.2%, -5.03%, 23.2%, and 26.5%, respectively, using B20. When B50 was used, the reductions were -14.5%, 4.5%, 21.4%, and 38.1%, respectively. Compared with B0, the reductions of total-BaP_{eq} emission factors were 64.2%, 49.8%, 57.2%, and 64.6%, respectively, using B10; 74.8%, 69.0%, 66.8%, and 71.0%, respectively, using B20; and 64.7%, 81.3%, 73.6%, and 85.9%, respectively, using B50. Notably, reducing the total-BaP_{eq} emission factor was in positive correlation with the added ratio of soybean biodiesel to petroleum diesel, except for using B20 at 0 loading (unload). This finding suggests that using biodiesel with soybean oil methyl ester can significantly reduce the BaP_{eq} of PAHs emitted from the generator.

3.4. Brake specific fuel consumption and energy efficiency

Table 3 lists the brake specific fuel consumptions (BSFCs) and energy efficiencies of generator under various added ratios of biodiesel to petroleum diesel. Again, the generator BSFC was positively correlated with the ratio of soybean biodiesel to petroleum diesel, regardless of loading. The BSFC decreased with an increasing load (in a negative correlation) despite of bio/petroleum diesel ratio. Because the heating value (9491.9 cal g⁻¹) of soybean biodiesel was lower than that (11035.7 cal g⁻¹) of pure petroleum diesel [60] by 1543.8 cal g⁻¹ (approximately 14.0%), the generator should consume more fuel using soybean biodiesel than when using petroleum diesel in order to maintain the same output. Consequently, the BSFC value of soybean biodiesel increased with an increasing bio/petroleum diesel ratio.

Energy efficiency (EE) refers to the ratio of the output energy divided by the input energy. The EE value increased with an increasing load (a positive correlation), regardless of bio/petroleum diesel ratio (Table 3). Lin et al. [33] indicated that although the use of

Table 3

The brake specific fuel consumptions and energy efficiencies of generator for the different fuel blends.

	5 kW		7 kW		10 kW					
	Avg.	Std.	Avg.	Std.	Avg.	Std.				
BSFCs (L kWh ⁻¹) ($n=3$)										
BO	0.480	0.001	0.405	0.0004	0.375	0.002				
B10	0.484	0.002	0.409	0.001	0.378	0.0003				
B20	0.488	0.001	0.411	0.006	0.380	0.0003				
B50	0.504	0.002	0.428	0.002	0.399	0.006				
Energy efficiencies (%) (<i>n</i> = 3)										
BO	19.53	0.05	23.15	0.02	25.04	0.11				
B10	19.57	0.06	23.18	0.05	25.09	0.02				
B20	19.59	0.02	23.24	0.33	25.12	0.02				
B50	19.45	0.08	22.90	0.09	24.59	0.35				

palm-biodiesel (bio/petroleum diesel ratio = 10% and 20%) could enhance the energy efficiency of diesel engine, the fuel blends with bio/petroleum diesel ratios from 20% to 100% resulted in incomplete combustion and obstruction of energy release, subsequently reducing the energy efficiency.

4. Conclusions

Among the diesel blends (B0, B10, B20, and B50) tested in this study, B20 exhibited the lowest PM emission concentration despite of the load (except for the 5 kW case), whereas B10 displayed lower PM emission factors when operated at 0 and 10 kW than the other fuel blends. Additionally, the emission concentrations of EC, OC, and TC were the lowest when B20 was used (with 14%, 29%, and 20% mean decreases, respectively, in comparison with those of B0) while B50 had the highest EC, OC, and TC emission concentrations, regardless of the loadings. Moreover, the mean TC content emitted from the generator ranged from \sim 39% to 62% and \sim 14% to 37%, respectively. In comparison with B0, the use of B10 or B20 could reduce EC, OC, and TC emission factors from the diesel generator by 2–41% (an average of 16%), 6–48% (an average of 27%), and 8–42% (an average of 20%), respectively, regardless of the loading.

PAHs emitted from the generator were mainly LMW-PAHs (an average of 76%). Under all evaluated loading, the average concentrations of total-PAHs emitted from the generator using the B10 and B20 were lower (by 38% and 28%, respectively) than those using pure petroleum diesel fuel (B0). Compared with B0, the emission factors of Total-PAHs decreased (an average of 72%) with an increasing ratio of biodiesel to premium diesel. Furthermore, using biodiesel with soybean oil methyl ester could significantly reduce the BaP_{eq} of PAHs emitted from the generator. Although generator BSFC was positively correlated with the ratio of soybean biodiesel to petroleum diesel, regardless of the loading, BSFC decreased with an increasing load despite the bio/petroleum diesel ratio. Adding soybean biodiesel into petroleum diesel increase recorded when using B20 (an average increase of 0.34%).

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